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**Description**

**Process for Liquefying a Hydrocarbon-Rich Flow While Simultaneously Recovering**  
**a C<sub>3</sub>/C<sub>4</sub>-Rich Fraction**

The invention relates to a process for liquefying a hydrocarbon-rich flow, in particular a flow of natural gas, with simultaneous recovery of a C<sub>3</sub>/C<sub>4</sub>-rich fraction, whereby the liquefaction of the hydrocarbon-rich flow is carried out in heat exchange for at least one refrigerant and/or mixed refrigerant flow, and the hydrocarbon-rich flow that is to be liquefied, after precooling, is subjected to a rectifying column, in which higher hydrocarbons are separated from the hydrocarbon-rich flow that is to be liquefied, and then is subjected to further cooling and liquefaction, whereby a C<sub>2+</sub>-rich fraction that is recovered in the subsequent cooling of the hydrocarbon-rich flow is fed to the rectifying column as a reflux liquid.

Processes of this type that are carried out in particular in natural-gas liquefaction plants are designed either as so-called LNG Baseload Plants – i.e., plants for liquefying natural gas for supply with natural gas as a primary energy – or as so-called Peak Shaving Plants – i.e., plants for liquefying natural gas to cover peak demand.

The above-mentioned Peak Shaving Plants are operated with turboexpanders or refrigerant mixtures in refrigeration circuits. The refrigeration circuits often contain only one or a few components.

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In these plants, the heavy hydrocarbons that are contained in the natural gas must be separated to produce the inventory of the mixture circuits, to cover losses during operation and because of product requirements.

This usually happens in that the hydrocarbon-rich flow that is to be liquefied is pre-cooled to a temperature of  $-10$  to  $-25^{\circ}\text{C}$  and then is fed to a rectifying column. By means of a suitable column configuration, the higher hydrocarbons – here, the  $\text{C}_{3+}$ -rich hydrocarbons as well as benzene are meant – are then separated from the flow to be liquefied in the rectifying column. At the top of the rectifying column, a  $\text{C}_2$ -rich fraction is drawn off and subjected to further cooling as well as liquefaction. In this case, the  $\text{C}_{3+}$ -hydrocarbon fraction that is recovered at the bottom of the rectifying column is concentrated with higher hydrocarbons to the extent that the rectifying column is set to the desired heating value in the  $\text{C}_2$ -rich top product.

The fraction that contains higher hydrocarbons and that is separated from the flow that is to be liquefied in the rectifying column is normally then separated into its components by rectification, whereby the latter are partially used as so-called Make-up Fractions - for example  $\text{C}_2\text{H}_6$  or  $\text{C}_3\text{H}_8$  – for the mixture circuit or circuits or are recovered as additional product flows and optionally are fed to a further processing.

The separation of the above-mentioned  $\text{C}_{3+}$ -rich fraction is performed with, for example, the following rectifying columns: demethanizer, de-ethanizer, depropanizer and optionally debutanizer. These columns allow the production of the following

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fractions: methane, ethane and ethane make-up, propane and propane make-up, LPG (liquid gas) as well as a  $C_{5+}$ -fraction.

It is disadvantageous in the above-described process, however, that the yield of  $C_4$ -hydrocarbons is comparatively low.

A generic process, in which in addition to the recycled  $C_{2+}$ -rich fraction, a  $C_4$  fraction is fed to the rectifying column as an additional reflux liquid, is known from the technical article "LPG – Recovery Processes for Baseload LNG Plants Examined" from OIL AND GAS JOURNAL (November 1997). This process allows a comparatively high  $C_3$  yield, but the expense as far as processing is concerned for the production of the  $C_4$  fraction that is required as a reflux liquid is significant.

The object of this invention is to indicate a generic process that makes possible the recovery of a  $C_3/C_4$ -rich fraction – the so-called LPG fraction – with as high a  $C_3$  yield as possible.

To achieve this object, it is proposed that a  $C_4/C_5$ -rich fraction be fed to the rectifying column as an additional reflux liquid, whereby the feed point of the  $C_4/C_5$ -rich fraction is at the top of the feed point of the  $C_{2+}$ -rich fraction, and a mass transfer zone is provided between the feed point of the  $C_4/C_5$ -rich fraction and the feed point of the  $C_{2+}$ -rich fraction.

To produce the  $C_4/C_5$ -rich fraction that is required as an additional reflux liquid, two processes that can also be combined are now conceivable.

If the higher hydrocarbons that are recovered in the rectifying column are separated by rectification in several steps that are downstream from the rectifying column, whereby one of these steps comprises the feeding of the higher hydrocarbons

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into a depropanizer, the  $C_4/C_5$ -rich fraction that is fed to the rectifying column as an additional reflux liquid can be recovered in a depropanizer side column, to which a  $C_{4+}$ -rich fraction that is drawn off from the depropanizer is fed. If the separation of ... by rectification that is downstream from the rectifying column comprises ...

corresponding to an alternative, advantageous embodiment of the process according to the invention.

To produce the  $C_4/C_5$ -rich fraction that is required as an additional reflux liquid, two processes that can also be combined are now conceivable.

If the higher hydrocarbons that are recovered in the rectifying column are separated by rectification in several steps downstream from the rectifying column, whereby one of these steps comprises the feeding of the higher hydrocarbons into a depropanizer, the  $C_4/C_5$ -rich fraction that is fed to the rectifying column as an additional reflux liquid can be recovered in a depropanizer side column, to which a  $C_{4+}$ -rich fraction that is drawn off from the depropanizer is fed. If the separation of higher hydrocarbons by rectification downstream from the rectifying column comprises a debutanizer, the  $C_4/C_5$ -rich fraction that is required as an additional reflux liquid can also be produced in the latter.

The possibility also exists of recovering the  $C_4/C_5$ -rich fraction that is fed to the rectifying column as an additional reflux liquid in a side column of the rectifying column by a  $C_{4+}$ -rich fraction being fed to this side column from the rectifying column.

The process according to the invention as well as additional embodiments thereof that represent the subjects of the dependent claims are explained in more detail below based on the embodiments that are shown in Figures 1 and 2.

In this connection:

Figure 1: shows an embodiment of the process according to the invention, in which the  $C_4/C_5$ -rich fraction is recovered in a depropanizer side column

Figure 2: shows an embodiment of the process according to the invention, in which the  $C_4/C_5$ -rich fraction is recovered in a side column of the rectifying column.

According to the process that is shown in Figure 1, a precooled and – if necessary – pretreated flow of natural gas, which has a temperature of between  $-10$  and  $-25^{\circ}\text{C}$ , is fed via line 1 to rectifying column T1. The optionally necessary pretreatment steps, such as, for example, drying,  $\text{CO}_2$  removal, sulfur removal, etc., are not considered in more detail below; the standard processes are known to one skilled in the art. The flow of natural gas that is brought in via line 1 typically has a pressure of between 30 and 90 bar.

A  $C_2$ -rich fraction is drawn off at the top of rectifying column T1 via line 2, and it is cooled in heat exchanger E1 to a temperature of between  $-25$  and  $-55^{\circ}\text{C}$  and in this case partially condensed. The partially condensed flow is then fed via line 3 to a separator D.

According to an advantageous embodiment of the process according to the invention, the benzene content of the  $C_4/C_5$ -rich fraction that is fed to rectifying column T1 as an additional reflux liquid – on which still more detail is given below – is less than 500 ppm, preferably even less than 300 ppm. This has the result that the  $C_2$ -rich fraction that is drawn off at the top of rectifying column T1 via line 2 has a benzene content of less than 1 ppm. An undesirable freezing-out of the benzene in the liquefaction part downstream from rectifying column T1 can thus be effectively avoided.

A  $C_1$ -rich fraction is drawn off via line 4 at the top of separator D and subjected to further cooling and liquefaction, not shown in Figures 1 and 2.

A  $C_{2+}$ -rich fraction is drawn off via line 5 from the bottom of separator D and fed to rectifying column T1 in its upper area as a reflux liquid.

A mass transfer zone M is arranged above the feed point of the  $C_{2+}$ -rich fraction that is fed via line 5 to rectifying column T1. The latter typically has 3 to 10 additional floors; this corresponds to about 2 to 7 theoretical floors.

A  $C_{3+}$ -rich fraction is drawn off via line 6 from the bottom of rectifying column T1 and optionally subjected to additional process steps. In this connection, a partial flow of the  $C_{3+}$ -rich fraction that is drawn off from the bottom of rectifying column T1 is fed as a reboiler stream to rectifying column T1 via line 7, in which a heat exchanger E2 is arranged.

The normally multistage rectification of the  $C_{3+}$ -rich fraction that is drawn off from the bottom of rectifying column T1 via line 6 is also sufficiently known to one skilled in the art with its different process variants. In this connection, line area 6' that is shown in dotted lines stood for the most varied processes, thus, for example, the feeding of the  $C_{3+}$ -rich fraction, drawn off via line 6, in a demethanizer as well as a downstream de-ethanizer. A  $C_{3+}$ -hydrocarbon-rich fraction that is treated as always is fed to depropanizer T2 via line 6''.

A  $C_3$ -hydrocarbon product fraction is drawn off via line 9 at the top of depropanizer T2, and it is cooled in heat exchanger E3. A partial flow of this fraction is then fed as reflux via line 10 to depropanizer T2. Product flows with a high  $C_3$  purity can be drawn off in gaseous or liquid form via lines 22 and 23; the latter are used, for example, as  $C_3$  make-up for the refrigerant mixture circuits. The residual portion of the  $C_3/C_4$  product fraction that is drawn off at the top of depropanizer T2 via line 12 – the so-

called LPG product fraction – is mixed in; the thus combined fractions are then released via line 13 and optionally fed to further processing.

A  $C_{5+}$ -rich hydrocarbon fraction is drawn off via line 8 from the bottom of depropanizer T2, and if desired, it is also fed to further processing.

A  $C_{4+}$ -hydrocarbon fraction is removed in depropanizer T2 via side hood 14, and it is fed to side column T3. Side column T3 is used in particular to remove benzene to a large extent from the fed  $C_{4+}$ -hydrocarbon fraction. For this purpose, a  $C_4/C_5$ -rich fraction is drawn off at the top of column T3 via line 16, cooled in heat exchanger E4 and fed via line 17 to a branch point, at which a partial flow is fed as reflux via line 18 to column T3, while the residual portion of the  $C_4/C_5$ -rich fraction is fed via lines 19 and 20 to rectifying column T1 as an additional reflux liquid.

A  $C_{5+}$ -rich fraction is drawn off via line 15 from the bottom of side column T3 and fed to depropanizer T2 in its lower area.

The  $C_4/C_5$ -rich fraction that is fed to rectifying column T1 as an additional reflux liquid is sub-cooled in a heat exchanger E5 preferably to a temperature of between  $-30$  and  $-50^{\circ}\text{C}$ . The optimum starting temperature of the  $C_4/C_5$ -rich fraction that is fed to rectifying column T1 is determined essentially by the conditions within rectifying column T1.

Instead of indirectly feeding the  $C_4/C_5$ -rich fraction that is used as additional reflux liquid, the latter can also – completely or partially – be fed via line 21, shown in dashes and dots, to separator D. The (partial) flow that is fed to separator D was then fed from its bottom via line 5 with the  $C_{2+}$ -rich fraction of rectifying column T1.



In the embodiment of the process according to the invention that is depicted in Figure 2, the  $C_4/C_5$ -rich fraction that is required as an additional reflux liquid now is not recovered in a side column of depropanizer T2, but rather in a side column 14 that is associated with rectifying column T1.

To this end, a flow is removed from rectifying column T1 in its lower area via line 30, and it is fed to side column T4. A  $C_{5+}$ -rich fraction is drawn off via line 31 from the bottom of this side column T4, and fed to rectifying column T1.

A  $C_4/C_5$ -rich fraction is drawn off via line 32 at the top of side column T4 and cooled in heat exchanger E6; a partial flow of this fraction is fed as reflux via line 33 of side column T4. The residual flow of the  $C_4/C_5$ -rich fraction that is drawn off via line 32 from the top of side column T4 forms the additional reflux fraction for rectifying column T1 and is fed to the latter via lines 34 and 35.

In turn, a heat exchanger E7, which is used in the cooling-off or sub-cooling of the  $C_4/C_5$ -rich fraction, can be provided in line 34.

Also, with this approach, the  $C_4/C_5$ -rich fraction that is used as an additional reflux liquid can be fed to separator D either partially or completely via line 36 that is shown in dashes and dots.

It is to be noted that an approach is selected, in which the  $C_3$  content of the  $C_4/C_5$ -rich fraction that is fed to rectifying column T1 is as small as possible to be able to carry out a separation of the  $C_{3+}$  components with the highest possible yield in rectifying column T1. Also, the content of  $C_4$  components in the  $C_4/C_5$ -rich fraction that is fed to rectifying column T1 should be as small as possible not to impair the high yield of  $C_4$ -hydrocarbons already achieved in rectifying column T1. Consequently, the content of

$C_5$ -hydrocarbons should be selected as large as possible to produce an efficient separation of  $C_{3+}$ -hydrocarbons.

The LPG that is recovered by means of the process according to the invention is therefore of great economic value, since LPG can be liquefied at ambient temperature, has a high energy content and is readily transportable. Moreover, LPG can undergo combustion in a nonpolluting manner. The  $C_3$  yield of the process according to the invention is more than 60%; that of the LPG's is more than 90%.